

ATTRIBUTION OF TRENDS AND VARIABILITY IN SURFACE OZONE OVER THE UNITED STATES

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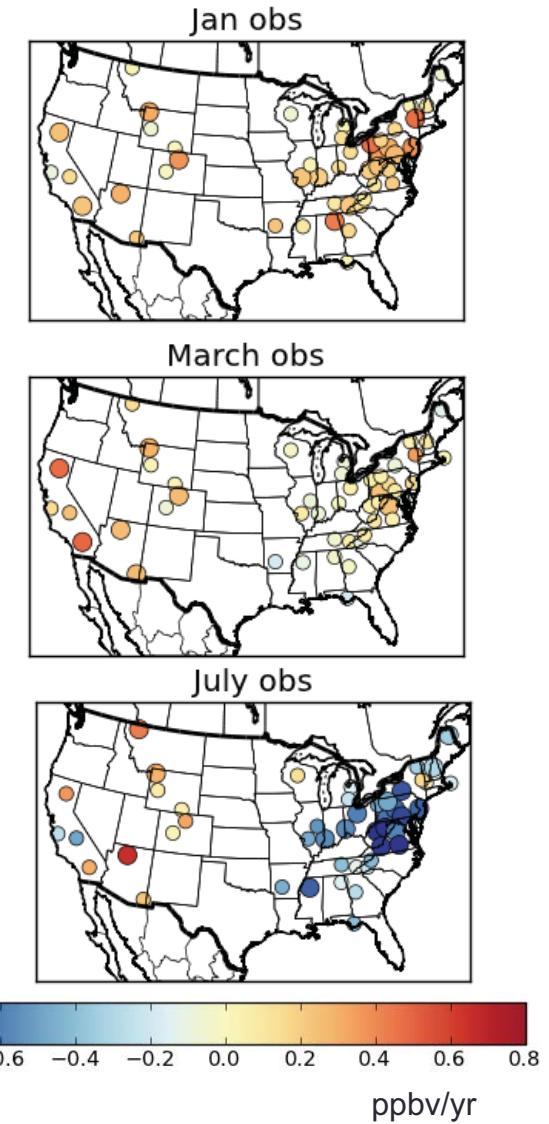
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Introduction

- CASTNET observations of surface O₃ from 1990-2010 [Cooper *et al.*, 2012] show:
 - negative trends in the eastern U.S. in summer, especially at the high end of the O₃ distribution; mixed trend directions in the western U.S.
 - Positive trends throughout US in winter & in western US in Spring
- Global model can reproduce E-W gradient in summer trends but not magnitude of western US trends [Koumoutsaris & Bey, 2012]
- Can a global model w/ interactive stratosphere reproduce the trends and inter-annual variability (IAV) in U.S. surface O₃ in different regions and seasons?
- How much do changes in emissions contribute to the trends? Strat-trop exchange?

Surface O₃ 1991-2010 trend



Model and Observations

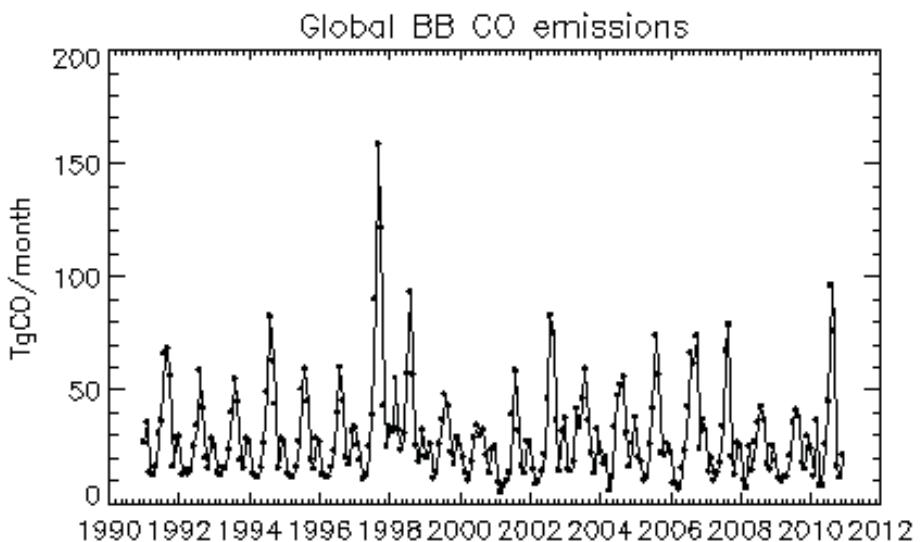
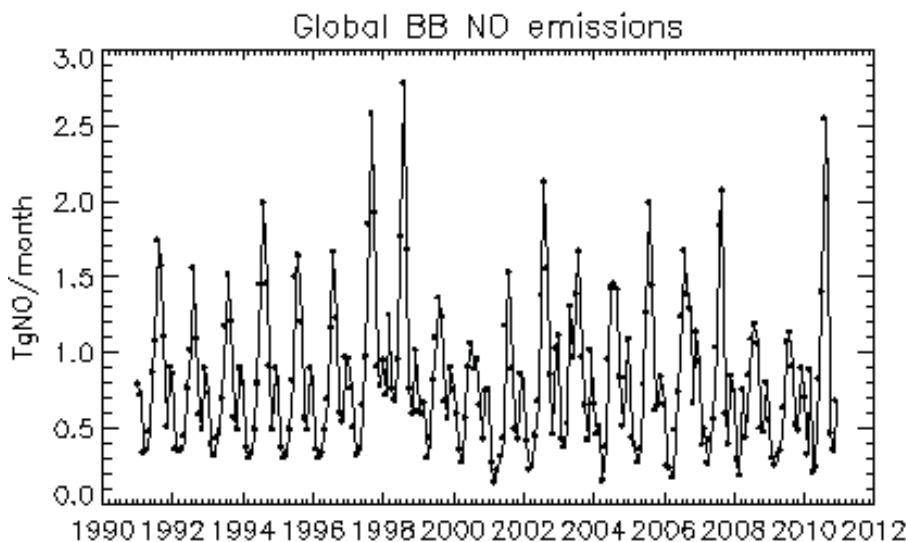
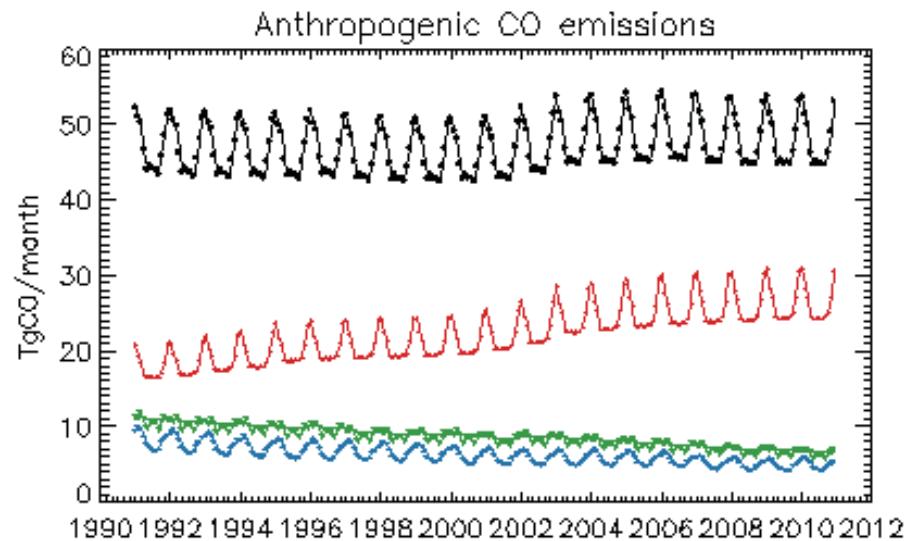
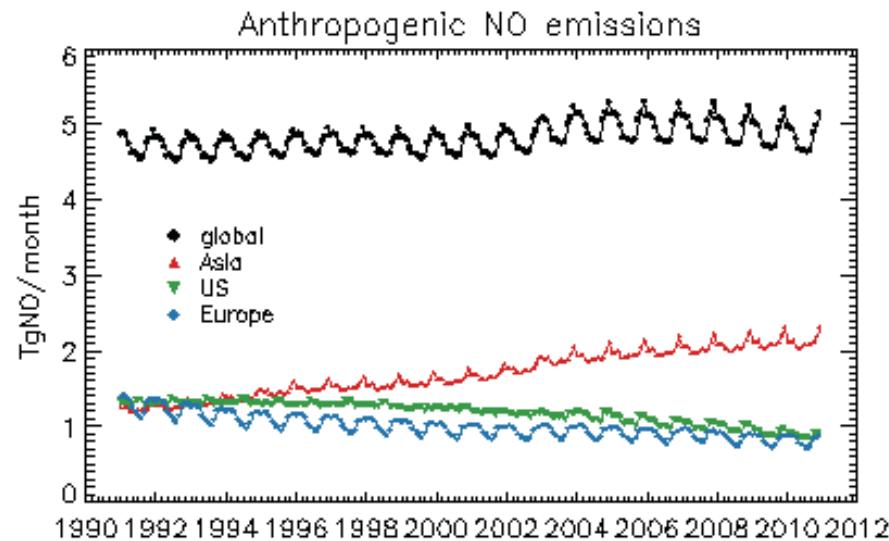
- Hourly observations from EPA Clean Air Status and Trends Network (CASTNET) rural surface stations, filtered as in *Cooper et al.* [2012]
- Mean, median, 5th & 95th percentile calculated for each month using all mid-day (11am-4pm) data
- GMI Chemical Transport Model (Duncan et al., 2007; Strahan et al., 2007) simulations of 1990-2010
 - Meteorology from the MERRA reanalysis
 - 2x2.5 degree horizontal resolution, 72 vertical levels
 - Stratospheric and tropospheric chemistry w/ 117 species, 400+ reactions
 - IGAC stratospheric SAD accounts for Pinatubo
 - Hourly output at station locations selected for mid-day hours

Emissions

- Standard simulation has time-dependent emissions
 - Biomass burning from GFED3 for 1997-2010; regional IAV based on *Duncan et al.* (2003) for 1990-1996
 - Anthropogenic emissions from EDGAR overwritten with NEI2005, EMEP, *Zhang et al.* (2009)
 - Annual scaling factors from GEOSChem for anthropogenic emissions (*van Donkelaar et al.*, 2008) for 1990-2006
 - 2007-2010 include annual scaling of whole U.S. based on NOx and CO emissions totals from EPA; REAS projections over Asia
 - 71% increase in Asian NO_x, 33% decrease in US NO_x for 1991-2010
- Sensitivity simulation with fixed emissions

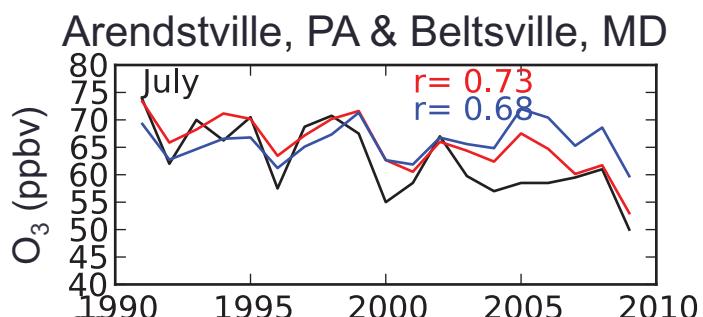
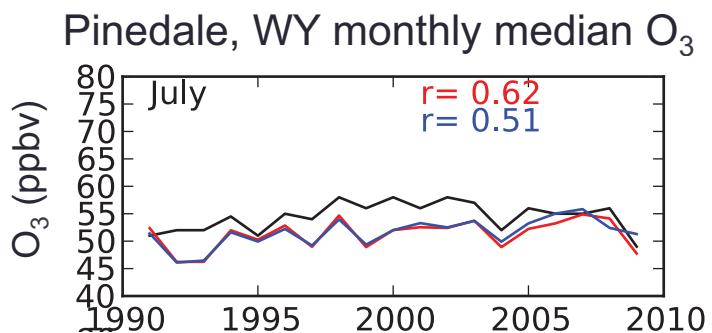
| Simulation | Anthro Emis | Biomass Burning |
|------------|----------------|-----------------|
| Standard | IAV | IAV |
| EmFix | Fixed at Y2000 | Fixed at Y2000 |

Emissions Time Series



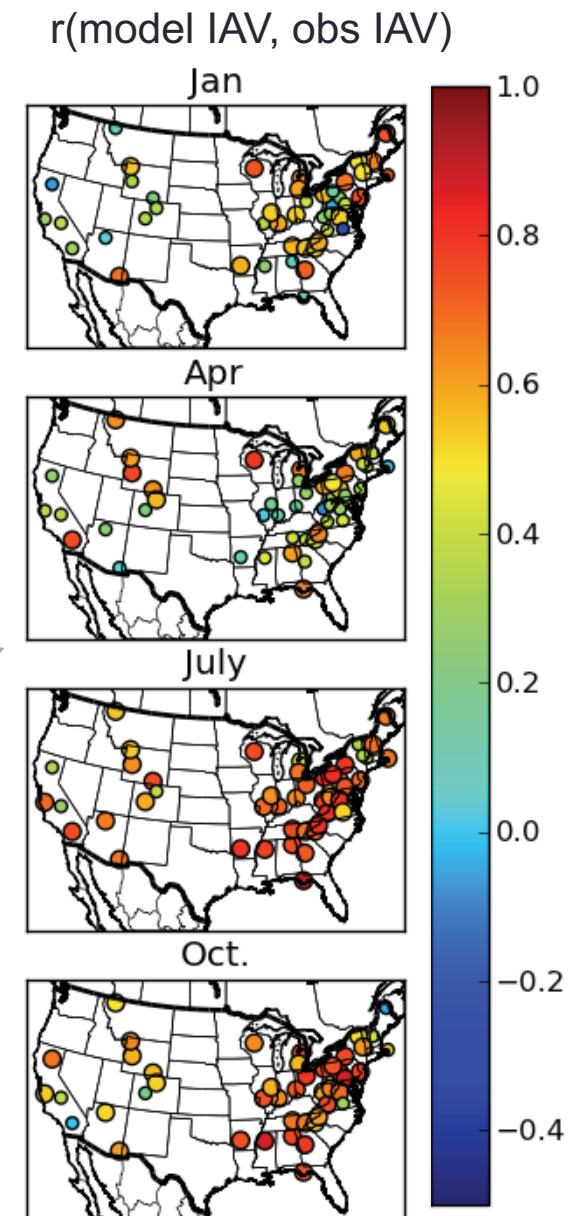
Modeled vs. Observed IAV

- Significant correlation of modeled and observed detrended IAV of median monthly O_3 at many stations
- Magnitude of variability underestimated
- Best correlations at eastern sites in summer, median or 95th percentiles
- Similar correlations in standard sim & EmFix



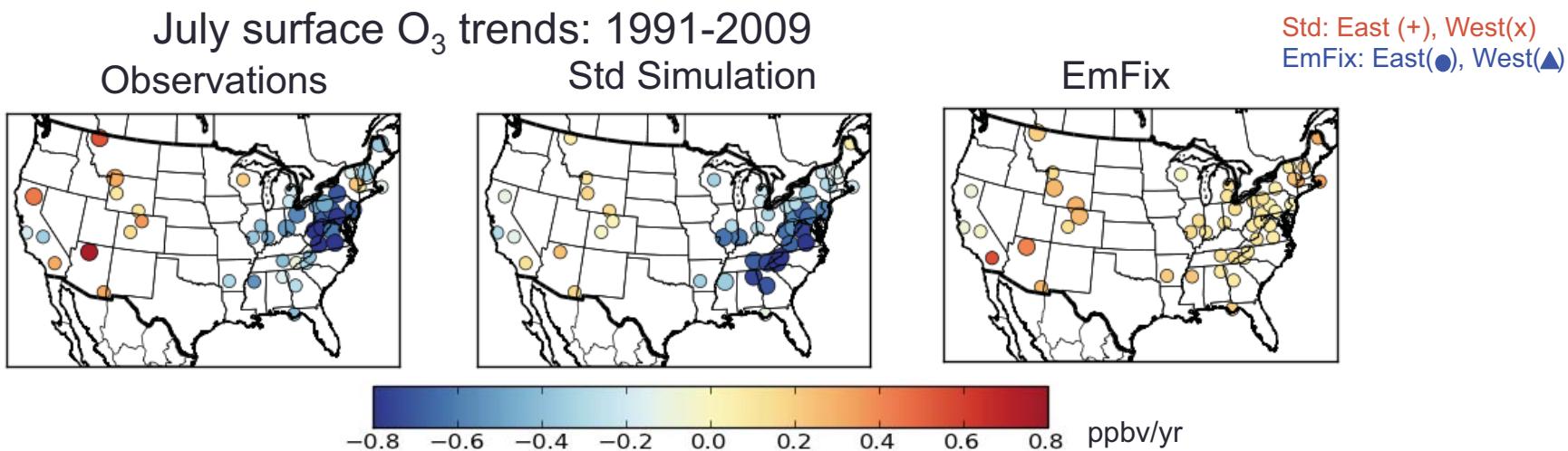
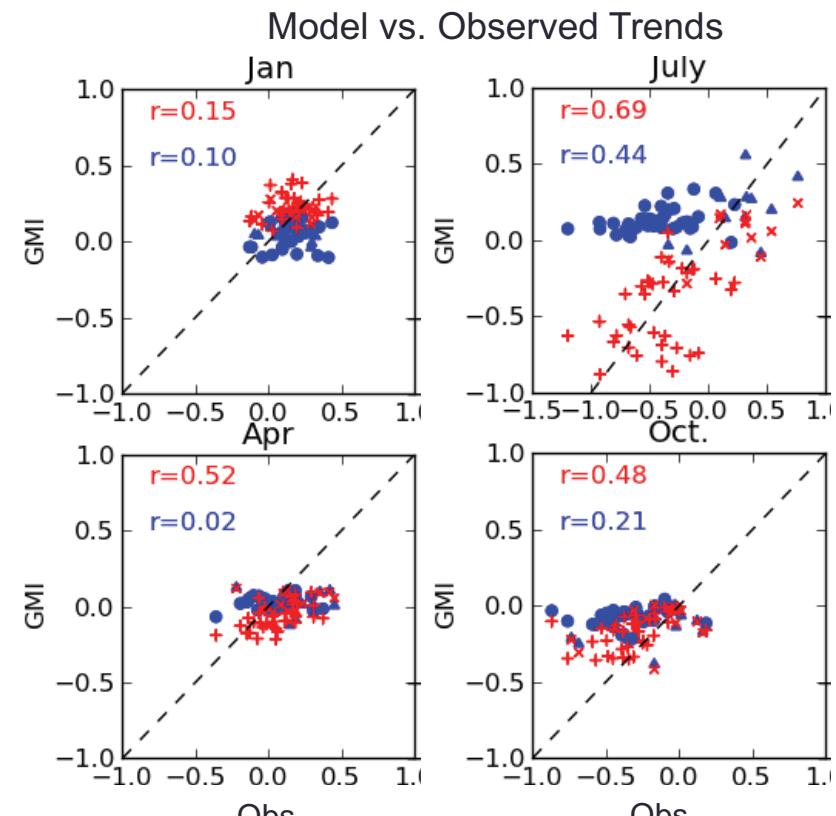
Observations
Standard (time-dependent emissions)
emFix (fixed emissions)

Large circles:
 r is significant
Small circles:
 r not significant



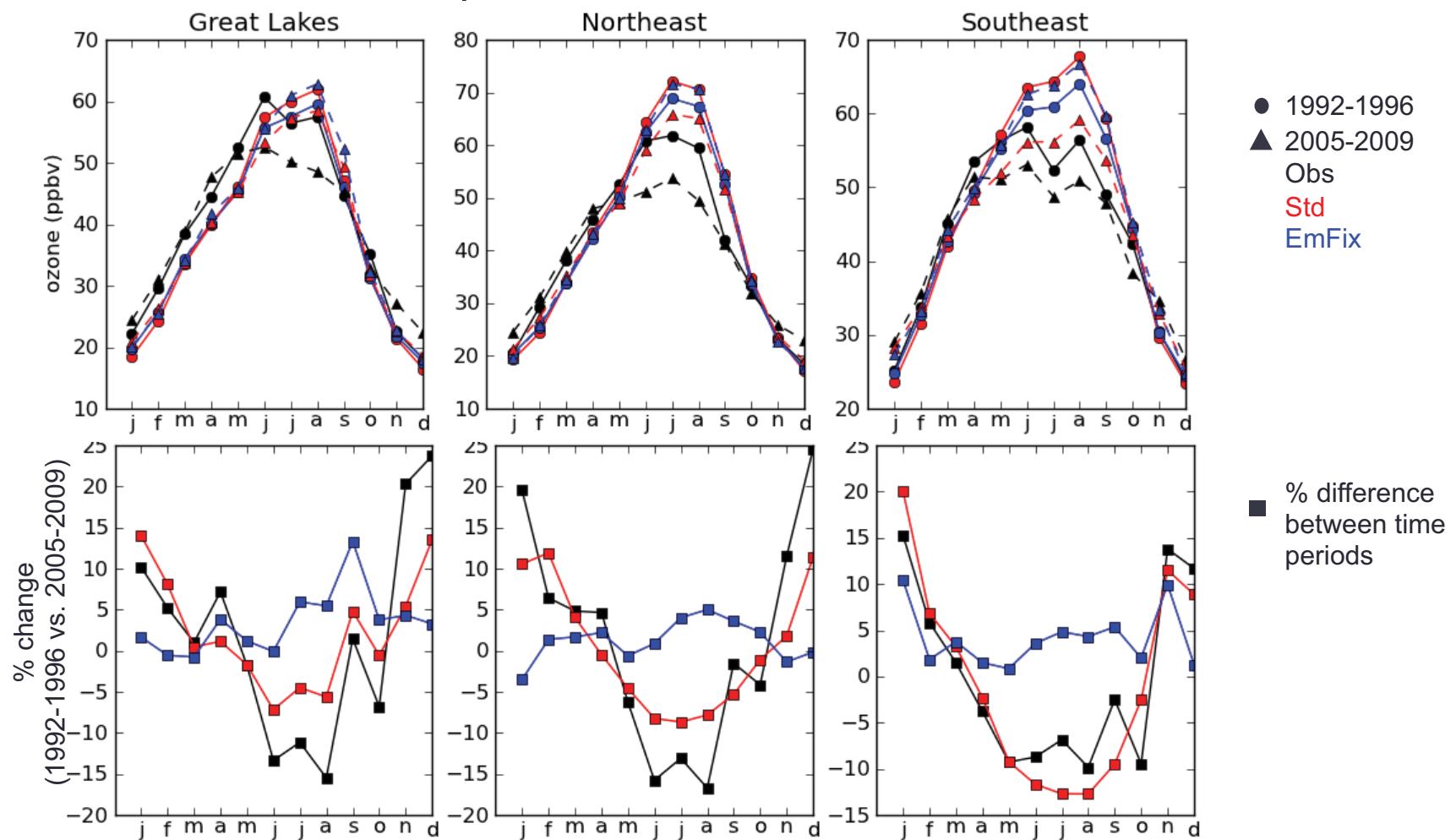
Summer Trends

- East-West gradient in summer trends captured by standard simulation but not EmFix simulation → key role for emission reductions
- Model underestimates magnitude of western trends



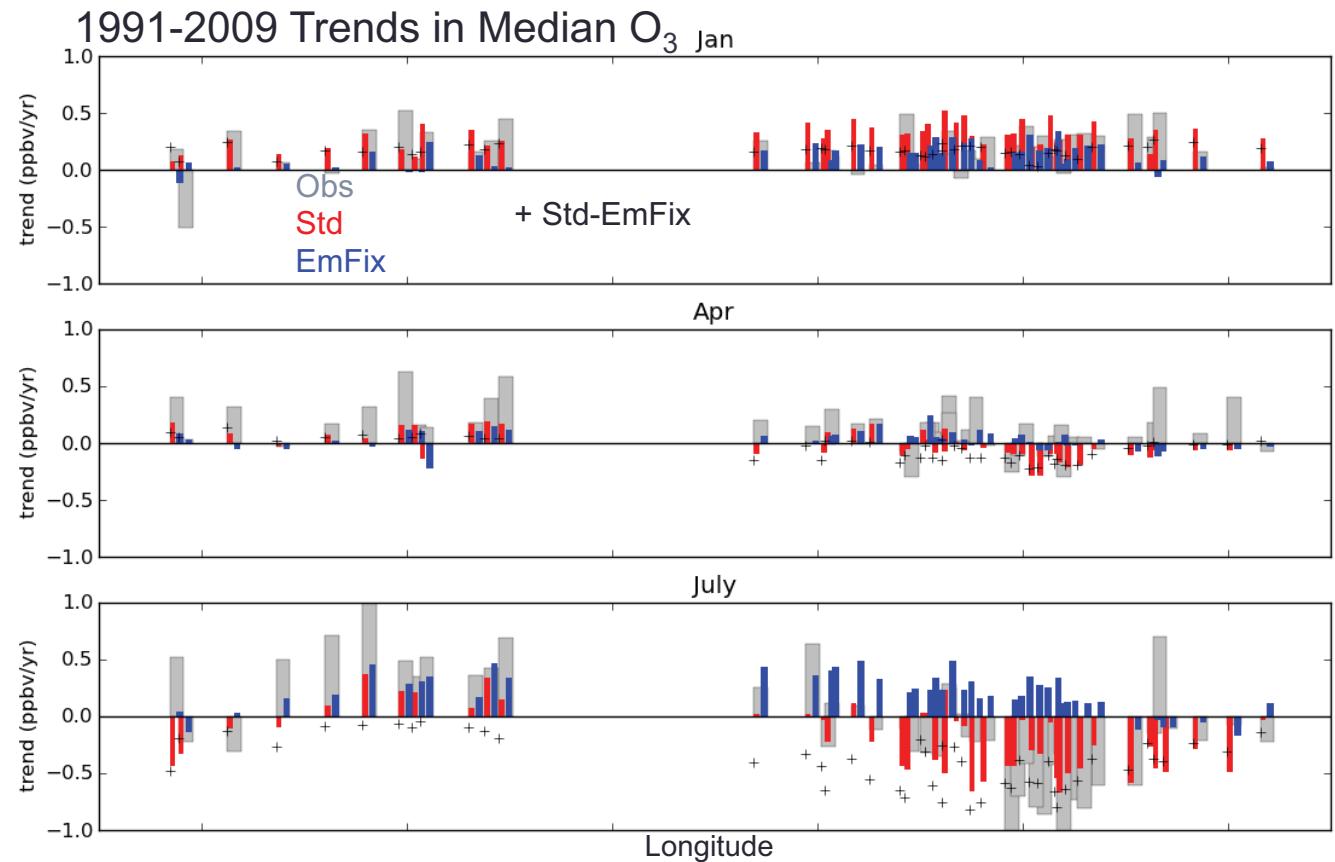
Shift in Seasonal Cycle

- Observed shift in seasonal cycle from summer peak to broader spring-summer max [Cooper et al., 2012]
- Standard simulation captures shift better than EmFix



Positive Trends in West and Winter

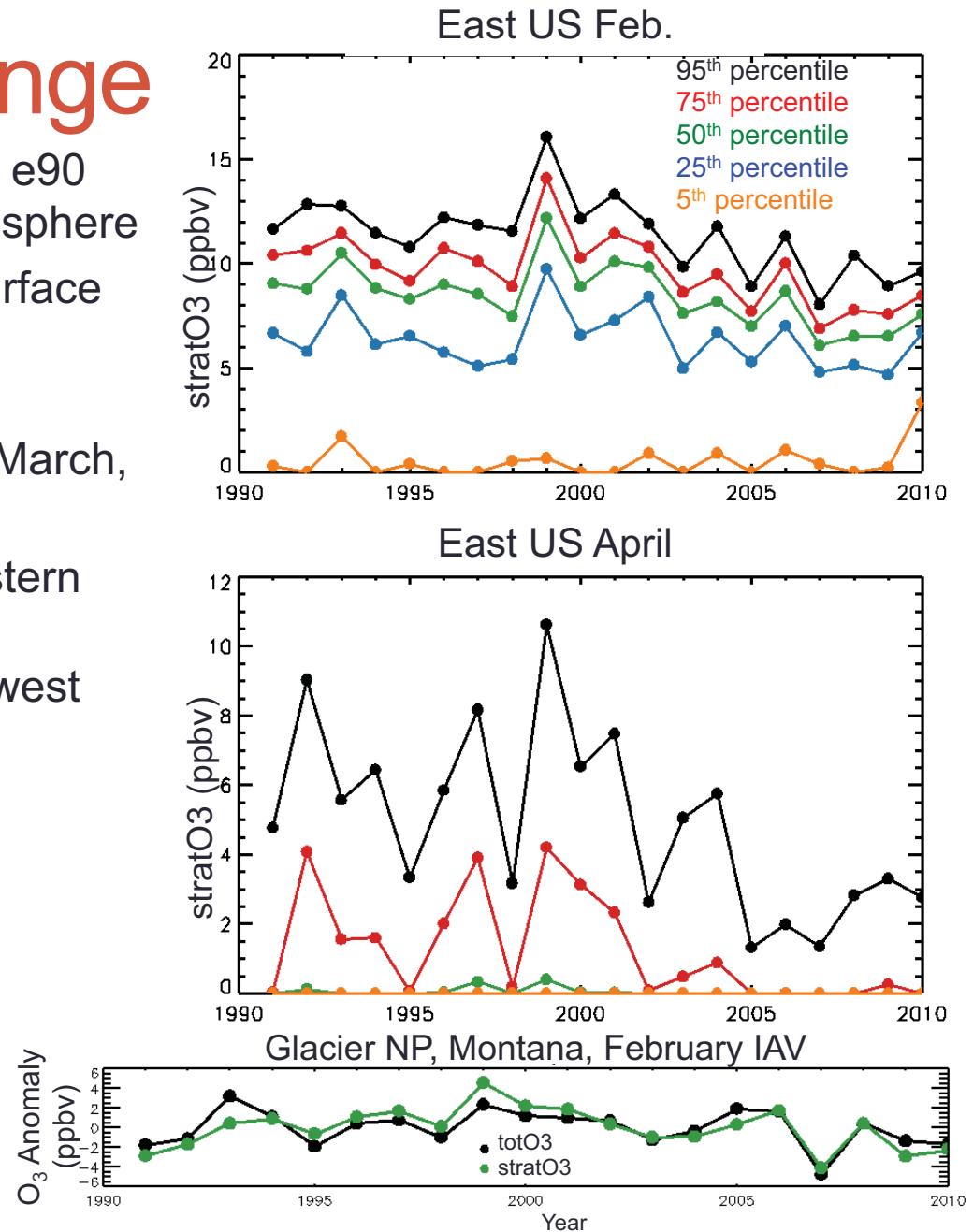
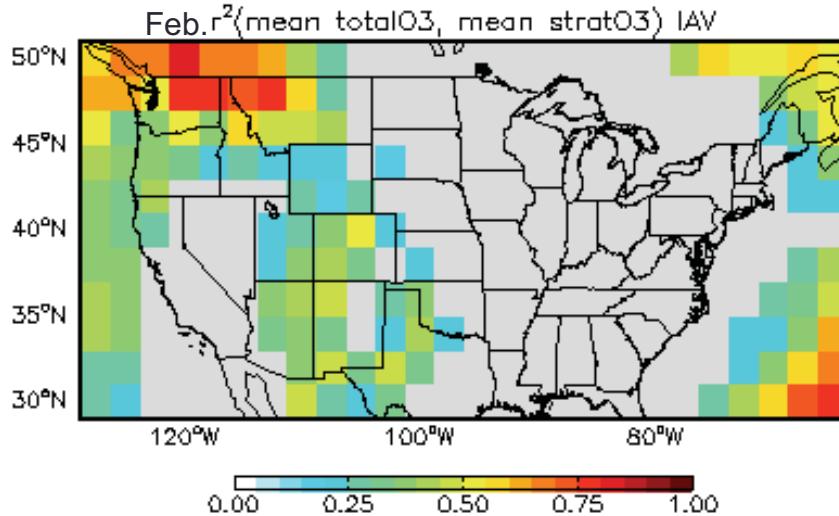
- Western trends more variable than eastern
- Positive trends due to changes in Asian, biomass burning, or local emissions?
Stratosphere-troposphere exchange (STE)?
Meteorology?



- Better agreement of Standard simulation in winter shows role for rising Asian emissions, while positive trends in EmFix in Summer show role for other factors
- Spring trends poorly captured → emission trends underestimated?

Strat-Trop Exchange

- Model stratO3 tracer equals O_3 above e90 tropopause, chemical loss in the troposphere
- No significant positive trends at the surface over the US for 1991-2010
- Significant negative trend in 50th-95th percentiles of east US stratO3 in Jan-March, 75th and 95th percentile in April
- Does not explain model's positive western US trends, but significant correlations between total & stratO3 winter IAV in west



Conclusions & Future Work

- 20-year hindcast captures east-west gradient in US summer ozone trends and the shift in seasonality toward a broader spring-summer max when time-dependent emissions are included
- Model underestimates positive trends in western US in spring and winter (and overestimates eastern trends in winter)
- Examined role of interactive stratosphere on modeled trends & IAV: No significant positive trend in monthly mean stratO₃ at surface; negative trend in high percentile surface stratO₃ over the eastern US

Future work:

- Conduct simulation with larger emission increase over Asia & spatially-varying emission changes over US based on satellite NO₂ (*Lamsal et al., 2011*)
- Quantify ozone trends on days with large Asian influence
- Examine impact of model resolution

